

First-order cancellation of the Cs clock frequency temperature-dependence in Ne-Ar buffer gas mixture

R. Boudot,¹ D. Miletic,² P. Dziuban,¹ C. Affolderbach,² P. Knapkiewicz,³ J. Dziuban,³ G. Mileti,² V. Giordano,¹ and C. Gorecki¹

¹FEMTO-ST Institute, CNRS, 32 avenue de l'observatoire 25044 Besancon cedex, France

²Time and Frequency Laboratory (LTF), Physics Department, University of Neuchâtel, Avenue de Bellevaux 51, 2000 Neuchâtel, Switzerland

³Wroclaw University of Technology, Janiszewskiego 11-17 Street, 50-372 Wroclaw, Poland

*rodolphe.boudot@femto-st.fr

Abstract: Through the detection of Coherent Population Trapping (CPT) resonances, we demonstrate the temperature-dependence cancellation of the Cs clock frequency in microfabricated vapor cells filled with a mixture of Ne and Ar. The inversion temperature at which the Cs clock frequency temperature sensitivity is greatly reduced only depends on the partial pressure of buffer gases and is measured to be lower than 80°C as expected with simple theoretical calculations. These results are important for the development of state-of-the-art Cs vapor cell clocks with improved long-term frequency stability.

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1. Introduction

A key point towards the development of vapor cell atomic frequency standards with improved long-term frequency stability is to reduce or even cancel the sensitivity of the clock frequency to temperature fluctuations [1]. In most of such frequency references, the alkali vapor cell is filled with a buffer gas or mixture of buffer gases. The presence of buffer gas causes three main phenomena: a reduction of the resonance linewidth through the Lamb-Dicke effect by reducing the free mean-path of alkali atoms [2], a homogeneous broadening and red shift of optical transitions [3,4] and a temperature-dependent shift of the clock frequency due to slightly dephasing interactions between alkali and buffer gas atoms or molecules [5]. The latter point explains the necessity to fill the cell with an optimized content to improve the clock performances.

The pressure and temperature shift coefficients of several pure buffer gases have been published in the literature. Nevertheless, reported values by different authors often suffer from discrepancy and lack of accuracy. This key problem makes difficult the realization of vapor cells with predetermined characteristics. Using available data, it has been demonstrated that a strong quadratic dependence of the Rb clock frequency on temperature can be obtained by using two buffer gases shifting each the clock transition in opposite directions. This technique is well-known towards the development of optically pumped passive rubidium frequency standards [6].

In the case of the Cs atom, most of the literature only reports measurements of linear coefficients due to a lack of resolution of frequency measurements in the experimental setup or to a limited temperature range studied [7, 8]. Recently, O. Kozlova et al. established first measurements of the quadratic coefficient for various buffer gases (Ne and N₂) [9] thanks to a high-resolution CPT Cs clock combining an original excitation scheme, a temporal Ramsey interrogation [10, 11] and allowing a great reduction of the light shift sensitivity [12]. At the same time and particularly, it has been measured in microcells [13] and cm-scale cells [9] that

the Cs clock transition frequency shows a quadratic dependence on temperature around an inversion temperature of about 80°C using only Ne as a unique buffer gas. In this case, it has been shown experimentally that the inversion temperature does not depend on the buffer gas pressure [14] as expected theoretically. This result could be of great interest for the development of simple configuration Cs CPT chip scale atomic clocks (CSAC) [15, 16] operating typically in this temperature range.

In the frame of the challenging european MAC-TFC project aiming to develop the first european CSAC prototype, Cs vapor microcells filled with a mixture of buffer gases (Ne and Ar) were realized in Wroclaw University as described in [17]. Mixtures with N₂ were not allowed due to the absorption of this gas by the Cs dispenser used for Cs activation and filling process [18]. Several microcells have been characterized in FEMTO-ST and UniNe laboratories using similar laboratory-prototype Cs CPT clocks.

In this article, we mainly investigate the temperature-dependence of the Cs clock frequency in vapor microcells filled with a mixture of Ne and Ar. A basic theoretical study based on temperature coefficients reported in [9] is given to describe the expected change of Cs frequency with temperature in such cells. Experimental tests based on a Cs CPT clock demonstrate the possibility to obtain a quadratic dependence on temperature of the Cs clock frequency in cells filled with a mixture of Ne and Ar. While quadratic dependence on temperature of the Cs frequency with N₂ and Ar buffer gas mixture has already been reported [20], this is to our knowledge the first reported observation of the temperature-dependence cancellation of the Cs clock frequency in cells containing a mixture of Ne and Ar.

2. Basic theoretical estimations

In vapor cell frequency standards, main phenomena that can shift the clock frequency are the Zeeman shift, the light shift and the buffer gas pressure shift. The frequency ν of the Cs hyperfine transition $|F = 3, m_F = 0\rangle \rightarrow |F = 4, m_F = 0\rangle$ can be described by:

$$\nu = \nu_0 + \alpha B^2 + \eta I(\omega) + \Delta\nu_{bg} \quad (1)$$

where ν_0 is the unperturbed Cs atom frequency (9.192 631 770 GHz), α is the second-order magnetic field coefficient (42.7 kHz/(mT)²), B is the static magnetic field, η is the light shift coefficient and $I(\omega)$ is the intensity of the pumping light. $\Delta\nu_{bg}$ is often commonly called the buffer gas pressure shift and is the major concern of this study. Assuming the contribution of light shift negligible and taking into account the Zeeman frequency shift, the pressure and temperature frequency shifts can be determined experimentally by measuring the change of the clock frequency as a function of the cell temperature. For a given buffer gas, the pressure shift of the clock frequency $\Delta\nu_{bg} = \nu - \nu_0$ is generally described in a wide temperature range by [5]:

$$\Delta\nu_{bg} = P[(\beta + \delta(T - T_0) + \gamma(T - T_0)^2)] \quad (2)$$

where P is the total buffer gas pressure at the reference temperature $T_0 = 273K$, T is the cell temperature, β is the pressure coefficient, δ is the linear temperature coefficient and γ is the quadratic temperature coefficient. Note that it can be useful sometimes to write P such as $P = P_r \frac{T_0}{T_r}$ to include directly in calculations the filling temperature T_r and the filling pressure P_r .

When a mixture of two buffer gases 1 and 2 is used, the Eq. (2) becomes:

$$\Delta\nu_{bg} = P[(r_1\beta_1 + r_2\beta_2) + (r_1\delta_1 + r_2\delta_2)(T - T_0) + (r_1\gamma_1 + r_2\gamma_2)(T - T_0)^2] \quad (3)$$

where β_i , δ_i and γ_i with $i = 1, 2$ are pressure and temperature coefficients for buffer gas 1 and 2 respectively. r_1 and r_2 denote the ratio between the buffer gas pressure and total pressure for the buffer gas 1 and buffer gas 2 respectively such as $r_1 + r_2 = 1$.

Another way to study cells filled with a mixture of two buffers gases is to define the ratio a between the pressure of buffer gas 2 and buffer gas 1 such as:

$$a = P_2/P_1 \quad (4)$$

This yields to the definition of global equivalent pressure and temperature coefficients such as:

$$\beta = \frac{\beta_1 + \beta_2 a}{1 + a} \quad (5)$$

$$\delta = \frac{\delta_1 + \delta_2 a}{1 + a} \quad (6)$$

$$\gamma = \frac{\gamma_1 + \gamma_2 a}{1 + a} \quad (7)$$

Substituting expressions from Eqs. (5) to (7) in Eq. (2), we can write:

$$\frac{d\Delta\nu}{dT} = P \left[\frac{\delta_1 + \delta_2 a}{1 + a} + 2 \frac{\gamma_1 + \gamma_2 a}{1 + a} (T - T_0) \right] \quad (8)$$

Nulling the expression of Eq. (8), we find that the pressure ratio a required to obtain an inversion temperature at $T = T_i$ is given by:

$$a = - \frac{\delta_1 + 2\gamma_1(T_i - T_0)}{\delta_2 + 2\gamma_2(T_i - T_0)} \quad (9)$$

Using the same principle, the expression of the inversion temperature T_i for a given pressure ratio a is obtained by:

$$T_i = - \frac{\delta_1 + a\gamma_2}{2(\gamma_1 + a\gamma_2)} \quad (10)$$

Figure 1 shows the estimated frequency shift of the Cs frequency versus the cell temperature for two different Ne-Ar pressure ratio and a total pressure of 6.6 kPa. Coefficients published in [9] are used.

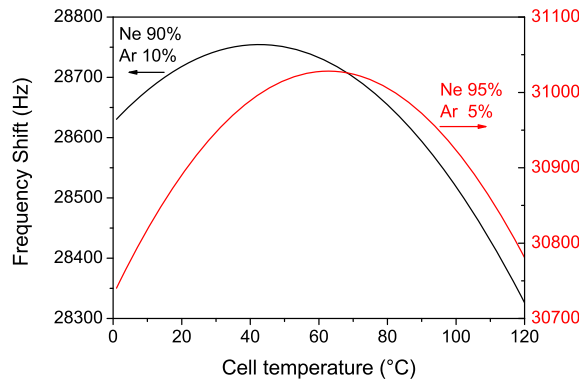


Fig. 1. Theoretical estimations from [9] of the Cs frequency shift versus the cell temperature for cells filled with a Ne-Ar mixture. The total pressure is fixed to be 6.6 kPa. The ratio are Ne-Ar (90%-10%) and (95%-15%) respectively.

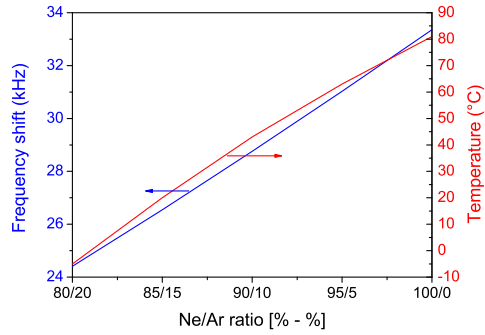


Fig. 2. Frequency shift and inversion temperature versus the Ne-Ar mixture for a Cs cell. The total pressure is 6.6 kPa.

Figure 2 shows the frequency shift of a Cs-Ne-Ar cell as well as the expected inversion temperature versus the Ne and Ar partial pressures in the cell. The total pressure is imposed to be 6.6 kPa.

It is clearly shown that there is no way to obtain any inversion temperature above about 80°C for Cs cells filled with a Ne-Ar mixture. For $a = 0$ (pure Ne cell), the temperature-dependence of the Cs frequency is expected to be cancelled at about 80°C as recently observed experimentally in chip-scale cells [13] and cm-scale glass blown cells [9]. Compared to pure Ne cells, the addition of Ar always shifts the inversion temperature to lower temperatures. The pressure ratio between Ar and Ne determines the inversion temperature. For example, the frequency of a Cs clock based on a vapor cell filled with a 85%Ne-15%Ar mixture is expected to exhibit a quadratic dependence on temperature around an inversion temperature of 20°C. This explains why the control of the filling pressure ratio and filling temperature is crucial when vapor cells with a mixture of buffer gases are realized. This issue is particularly critical in microfabricated cells because high temperatures are required to operate the cell sealing with anodic bonding. Such values can be of great interest for the development of vapor cell Cs frequency standards using cm-scale cells. The total frequency shift depends on both the pressure ratio and the total pressure and is increased with the quantity of Ne.

3. Experimental set-up

Two Cs CPT clocks have been implemented in FEMTO-ST and University of Neuchâtel (UniNe) laboratories respectively. Both clocks are quasi-identical and based on similar components. Figure 3 shows a typical schematic of the experimental Cs CPT clock.

The optical source is a 1.5 MHz-linewidth Distributed Feedback (DFB) laser resonant with the Cs D1 line at 894 nm. Two phase-coherent optical sidebands needed to perform the CPT spectroscopy are generated by modulating an external temperature-controlled pigtailed electro-optical phase modulator (EOM) at 9.192 GHz. At the output of the EOM, the laser intensity is stabilized using an acousto-optical modulator (AOM)-based power servo loop. For this purpose, a fraction of the laser power is extracted using a beam splitter cube and detected by the low noise photodiode PD2. The detector output voltage is compared to a high-stability reference voltage. An error signal is generated, processed into a simple analog PI controller and sent into the amplitude modulation (AM) input port of the synthesizer driving the AOM. This servo loop allows to reduce the relative laser intensity fluctuations at the output of the EOM by a factor 25 at 1000 s integration time. At the output of the AOM, the collimated laser beam is

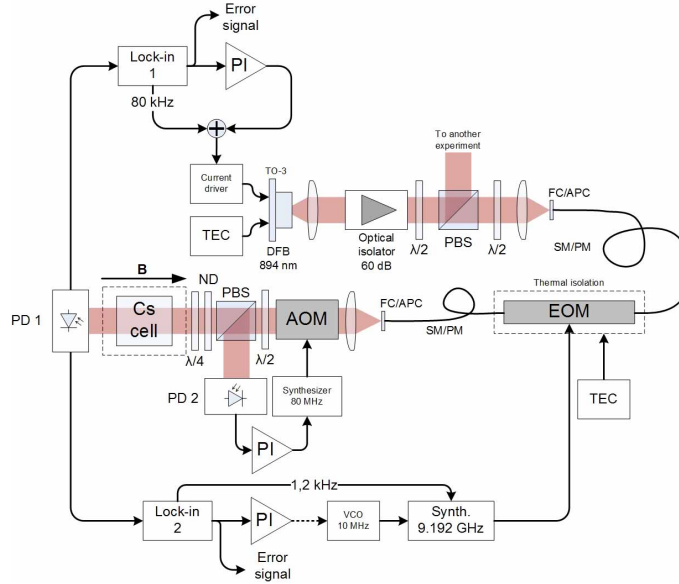


Fig. 3. Schematic of the Cs CPT clock used to test Ne-Ar microcells.

sent through a neutral density filter to attenuate the optical power and a quarter-wave plate to circularly polarize the beam.

CPT interaction occurs in a 2-mm diameter and 1.4-mm long microfabricated cell realized according to the process flow described in [17–19]. The cell temperature is stabilized at the mK level using a commercial temperature controller. The cell is surrounded by a solenoid to apply a static magnetic field of $10 \mu\text{T}$ and raise the Zeeman degeneracy. The ensemble is isolated from spurious magnetic fields using two concentric cylindrical mu-metal magnetic shields. The optical power transmitted through the cell is detected by the low noise Si photodiode PD1. Two servo loops are then implemented from the output voltage of the photodiode PD1. The first one is used to lock the laser frequency near the center of the homogeneously broadened absorption line by modulating the DFB laser current at 80 kHz and demodulating it with the lock-in amplifier 1. The second servo loop aims to lock the frequency of the 9.192 GHz signal driving the EOM on the Cs 0-0 hyperfine transition by modulating at 1.2 kHz the microwave synthesizer frequency. The lock-in amplifier 2 allows to increase significantly the signal to noise ratio of the detected CPT resonance. Once the 9.192 GHz signal locked on the atomic resonance, the clock frequency is measured by comparison with the signal from a microwave synthesizer referenced by a reference hydrogen maser.

The main difference between FEMTO-ST and UniNe clocks are the following. The microwave modulation frequency is 9.192 GHz and 4.596 GHz in FEMTO-ST and UniNe respectively. However, this does not change the physics of the buffer gas related temperature coefficient studied here. Also, in UniNe experiment, the DFB laser exhibits a linewidth of 15 MHz and no laser power servo loop is implemented.

4. Experimental results

Several microcells have been realized. Experimental results on four of them are reported here. All the cells are expected to be filled with 85 % of Ne and 15 % of Ar. Various total pressures are expected. Cells 1, 2 and 4 were tested in FEMTO-ST laboratory while the cell 3 was tested

in UniNe.

Figure 4 shows a typical CPT resonance observed in a microcell filled with a buffer gas mixture of Ne and Ar.

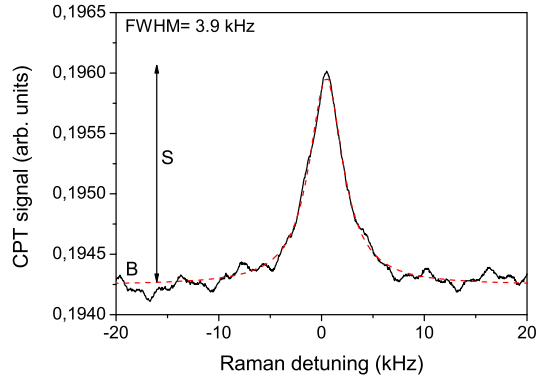


Fig. 4. Dark line resonance detected in a Cs-Ne/Ar microcell. The solid line is experimental data while the dashed line is a lorentzian fit. The laser power is $20 \mu\text{W}$. The cell temperature is 75°C . The total buffer gas pressure is measured to be 8.5 kPa.

The linewidth of the dark resonance is measured to be 3.9 kHz. The contrast of the CPT resonance, defined as the ratio between the height S of the dark resonance and the DC background voltage B , is measured to be about 0.9 %. According to theoretical expectations, we measured that the CPT resonance linewidth is found to be broader in a Ne-Ar microcell than in a pure Ne microcell for identical experimental configurations.

The buffer gas pressure shift measurement procedure is similar to the one described in [13]. The microcell temperature is changed. For each temperature, the clock frequency is measured for various laser intensities to determine the linear light shift function. The frequency shift is then estimated by extrapolation at zero intensity and subtraction of the Zeeman shift.

Figure 5 shows the frequency shift of the Cs 0-0 hyperfine resonance as a function of the temperature for different microcells.

The circles, stars, squares and triangles are the experimental data while solid lines are a computer fit of the Eq. (3) to the data. For the four cells, it is clearly shown that the temperature-dependence of the Cs clock frequency is canceled for inversion temperatures lower than 80°C as expected in Fig. 2. Data fits are respectively $24915 + 11.16T - 0.10T^2$, $36472 + 36.49T - 0.39T^2$, $36170 + 15.39T - 0.14T^2$ and $51823 + 29.10T - 0.47T^2$ for the cells 1, 2, 3 and 4 respectively. The inversion temperature is measured to be 51.6, 46.4, 53.0 and 31.7°C for the cell 1, 2, 3 and 4 respectively.

Table 1 resumes measured characteristics of the tested Cs-Ne-Ar microcells. All the presented cells have been realized separately in different runs.

For the different cells, the linewidth of the CPT resonance tends to be lower for increased buffer gas total pressure as expected for this buffer gas pressure range. In general, the measured total pressure is found to be about 2-3 times smaller than the expected one. We note also that the measured pressure ratio is not in correct agreement with the expected one. Further tests are in progress to control accurately the actual filling total and partial pressures. Nevertheless, experimental data (measured on different experimental Cs clocks) are in correct agreement for cells 2 and 3 expected to be filled with the same pressure of 24 kPa. This indicates a good repeatability of the cell fabrication process and that only a re-scaling of the apparatus is needed

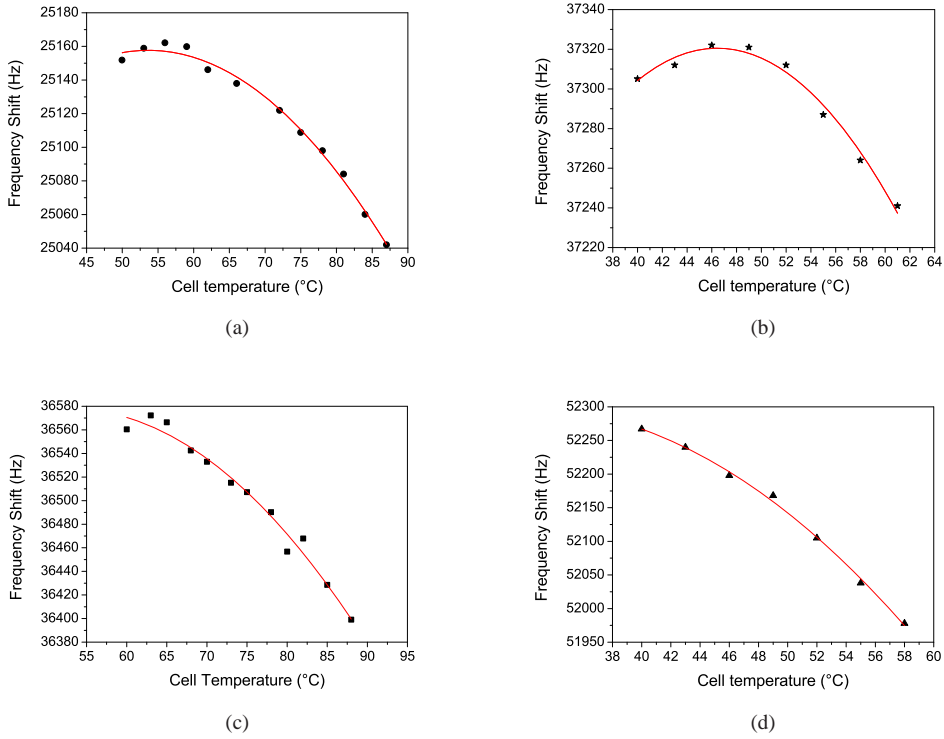


Fig. 5. Frequency shift of the Cs clock versus the cell temperature in microcells filled with a Ne-Ar mixture. The microwave power driving the EOM is 22 dBm. (a): Cell 1. (b): Cell 2. (c): Cell 3 (d): Cell 4.

Table 1. Measured characteristics of the tested Cs-Ne-Ar microcells. The actual measured total buffer gas pressure P_{meas} and Ne proportion (in %) in the cell is estimated using data fits reported above and pressure and temperature shift coefficients reported in [9]. The expected total buffer gas pressure P_{exp} is also reported to be compared to the measured one. The uncertainty on T_i and Δv_{bg} is given to be $\pm 2^\circ\text{C}$ and $\pm 10\text{ Hz}$ respectively. From these values and uncertainties of pressure shift coefficients reported in [9], we calculate that the uncertainty on the Ne proportion and P_{meas} is typically $\pm 3.4\%$ and $\pm 0.9\text{ kPa}$ respectively for each cell

Cell	FWHM (kHz)	T_i ($^\circ\text{C}$)	Δv_{bg} (Hz)	P_{meas} (kPa)	P_{exp} (kPa)	Ne proportion (%)
1	4.2	51.6	24915	5.6	15	92.4
2	3.9	46.4	36472	8.4	24	91.1
3	3.7	53.0	36170	8.1	24	92.7
4	3.2	31.7	51823	12.5	26	87.7

to obtain cells with desired characteristics.

5. Conclusions

We studied the frequency-temperature dependence of the Cs clock frequency in microcells filled with a mixture of Ne and Ar. Simple theoretical estimations have been done and can be extended to others buffer gas mixtures. It has been measured that a quadratic temperature dependence of the Cs frequency can be obtained with such microcells for inversion temperatures lower than 80°C. The inversion temperature only depends on the Ne-Ar pressure ratio and is shifted to higher temperatures for cells with increasing partial pressure of Ne. The total frequency shift depends on both the total and partial pressures. These properties highlight the necessity to control accurately the filling pressure and temperature during the cell filling process. The presented results are of high relevance for the development of any type of Cs vapor-cell frequency standards.

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